

PRODUCING TRANSMUTATION ELEMENT ON MULTI-LAYERED Pd SAMPLE BY DEUTERIUM PERMEATION

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Time-of-flight secondary ion mass spectroscopy (TOF-SIMS) was employed for elemental analysis of surface of multi-layered palladium sample with small amount of cesium. Substances with mass number 135 and 137 were newly detected by TOF-SIMS after deuterium permeation at 70°C. The result suggests production of an element with mass number 137 which would be produced from cesium by same low-energy nuclear reaction.

1. Introduction

The phenomena on nuclear reactions at low temperature in solid-state have been widely investigated for these 17 years. Among several experimental methods for the reaction, the gas permeation method is one of the promising methods. Iwamura *et al.*¹ have studied using this method with Pd film complexes, which consist of Pd layers, CaO layers and a bulk Pd. They have reported a low-energy nuclear reaction such as the transmutation of Cs into Pr in deuterium permeation experiment and have recently found a certain rule of nuclear transmutation, that is, 8 mass number and 4 atomic number increase in the process. The phenomenon has been observed with good reproducibility.

While, we have been taking account of the possibility of nuclear transmutation not only in such Pd film complexes but also in plain Pd foil by hydrogen gas permeation at room temperature. We have performed an elemental analysis on the Pd foil and have reported increases in count intensity of several elements including Ag after hydrogen permeation.² The count intensity of Fe has been sometimes observed to increase significantly after the permeation of highly pressurized hydrogen gas through Pd samples of 0.1 and 0.3 mm thickness. This result would show a pressure effect on the reaction of transmutation. Furthermore, the isotopic composition of Ti and Cr has been observed to differ from the natural isotopic abundance of those elements. These results have suggested that several elements were produced

by a nuclear transmutation and that the reaction could occur in hydrogen system as some researchers have claimed in various experiments.³⁻⁵

The aforementioned results suggest that there is a key to understand the reaction in terms of the mobility of proton or deuteron in Pd lattice. These reports caused us to study the phenomena systematically by the permeation experiment for deuterium as well as hydrogen using multi-layered Pd samples. Thus, in this present investigation, we have performed a deuterium permeation experiment using Pd samples⁶ including a multi-layered sample consisting of single couple of CaO and Pd thin films and a bulk Pd foil.

On the basis of the reports on transmutation of ^{133}Cs into ^{141}Pr , production of elements with mass number ranging between 133 and 141 could be expected before Pr production. Therefore, we have focused our attention to this mass number range to search for the producing elements as a result of low-energy nuclear reaction. The amount of newly producing elements in this range would be so little; it is desirable to employ an instrument having a good sensitivity for a very small quantity of the elements on the sample with high resolution in mass number. Consequently, we have performed the element analysis using time-of-flight secondary ion mass spectroscopy (TOF-SIMS).

2. Experimental

A base Pd foil (99.95% pure) of $0.1 \times 12.5 \times 12.5 \text{ mm}^3$ in size was rinsed with acetone and pure water, and then washed by aqua regia to remove impurities on the Pd foil surface. Next, the Pd was annealed at 900°C for 8 h, followed by being cooled to room temperature in furnace and washed again with aqua regia. Using the base Pd foil, we prepared three types of samples. The first one was a plain Pd sample just as the base Pd foil was. It had no deposition of any additional elements. The second one had small amount of Cs deposited on the base Pd foil. The last one had small amount of Cs on the uppermost of multi-layered Pd sample. The multi-layered sample consisted of a couple of CaO and Pd thin films on the base Pd foil. The CaO and Pd thin films were formed on the Pd foil by Ar ion beam sputtering under the input energy of 30 W for 5 min and 20 W for 1 min, respectively. After forming the thin films, small amount of Cs was deposited on the multi-layered Pd sample by an electrochemical method. In this method, the Cs was deposited by applying an electric field to 0.5 mM Cs_2CO_3 solution; a 1 V negative voltage was applied to the multi-layered Pd sample for 10 s. A Pt foil of $10 \times 10 \text{ mm}^2$ in size was utilized as a counter-electrode. The thickness of CaO and Pd films formed were 2 and 40 nm, respectively. No deuterium gas was loaded to the samples before deuterium permeation experiment.

Figure 1 shows the deuterium permeation system to investigate the transmutation of Cs into other elements of larger mass number. Whole the permeation system was constructed at Ulvac Techno, Ltd. in Japan. The surface inside of chamber was finished by electro-polishing method to reduce amount of impurity molecules deposited on the surface as low as possible. A stainless steel sample holder for

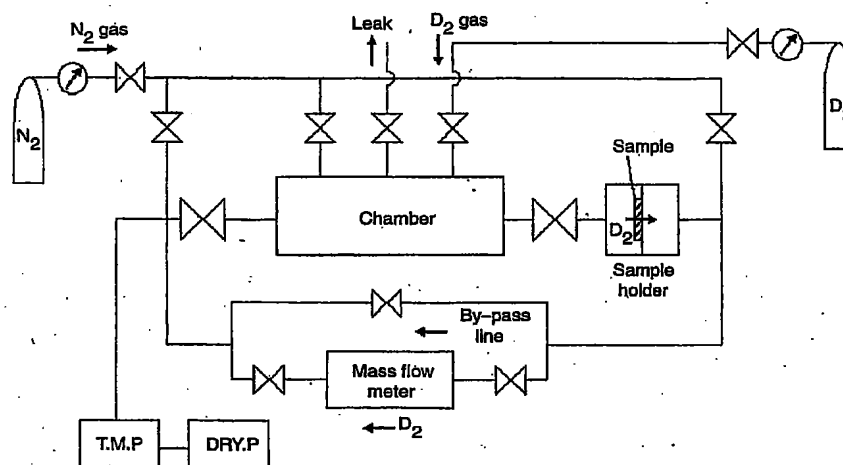


Figure 1. Apparatus for cold transmutation.

the deuterium permeation system is shown in Fig. 2. The vacuum chamber and the sample holder were baked at 200°C sufficiently before setting the sample. The chamber is usually filled with N_2 gas under non-experiment condition.

Just before the permeation experiment, the Pd samples were set into the sample holder in an air environment and it was placed at the vacuum chamber. Then, the chamber was filled with deuterium gas at a pressure 0.1 MPa; the thin Pd film side of multi-layered Pd sample was exposed to D_2 gas. The other side of

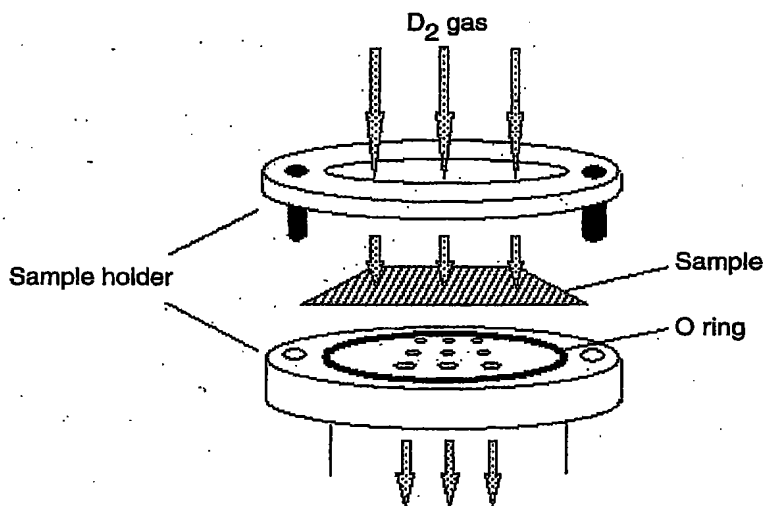


Figure 2. Sample holder for deuterium permeation.

sample was evacuated by a turbo molecular pump to prevent the Pd sample being contaminated from the atmosphere. The deuterium permeated from the chamber through the Pd sample to the evacuated side by the pressure gradient for about 1 month. A heater was employed to keep the temperature of the chamber at 70°C during the experiment. After the permeation experiment, the heater is turned off and the chamber was filled with N₂ gas, then the sample ("after permeation sample") was taken out from the holder. Before the element analysis, the sample was not treated for purging the deuterium atoms remaining in. The sample surface of gas-filled side was analyzed by TOF-SIMS (ULVAC-PHI: TFS-2100).

TOF-SIMS has a good sensitivity for a very small quantity of the elements on the sample with high resolution in mass number although it is difficult to deduce the absolute quantities from its output data alone. The primary ion in TOF-SIMS was Ga⁺ and we measured three randomly selected areas of 40 × 40 μm². The spectra, given in this paper, were obtained after sputter cleaning of upper most surfaces of samples by the Ga⁺ for 10 s. In order to take into account the contamination from the environment, we prepared the control sample ("control sample") without flowing the deuterium gas, which was prepared by the same procedure for the permeation samples. Comparing the composition of the elements on the surface of "Control sample" with that of "after permeation sample", or comparing that of plain Pd sample with that of multi-layered Pd sample, we tried to specify newly produced elements during the deuterium permeation.

3. Result and Discussion

A TOF-SIMS spectrum of mass number range 132–141 without deuterium permeation for Cs deposited on plain Pd foil is shown in Fig. 3. No marked

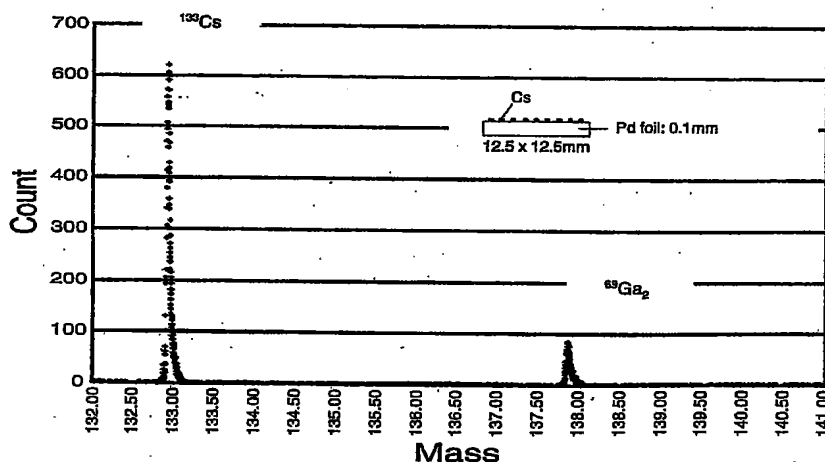


Figure 3. TOF-SIMS spectrum of mass number range 132–141 without deuterium permeation for Cs deposited on plain Pd foil.

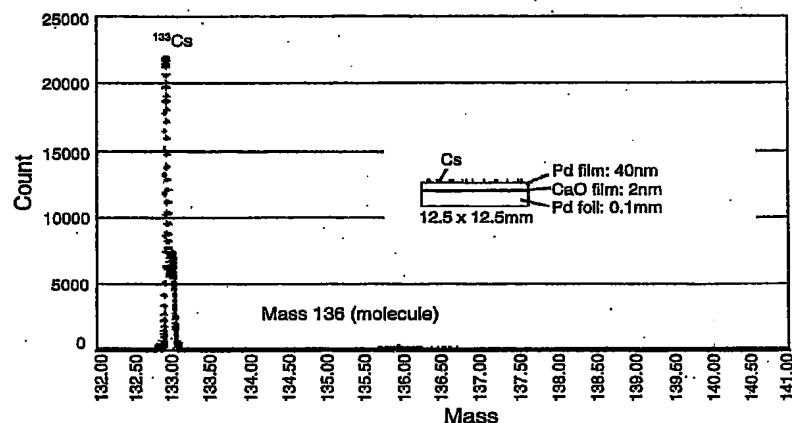


Figure 4. TOF-SIMS spectrum of mass number range 132–141 without deuterium permeation for Cs deposited on multi-layered Pd sample.

count, except ^{133}Cs and $^{69}\text{Ga}_2$ was seen in this spectrum for this control sample. A TOF-SIMS spectrum of mass number range 132–141 without deuterium permeation for Cs deposited on the multi-layered Pd sample is shown in Fig. 4. The sample was used for another control one and the schematic view of sample is shown in this figure. No marked count was again observed, even though only a few count due to a molecule contaminant at mass number 136 was detected. A TOF-SIMS spectrum of mass number range 132–141 after deuterium permeation at 70°C for the plain Pd foil is shown in Fig. 5, where no

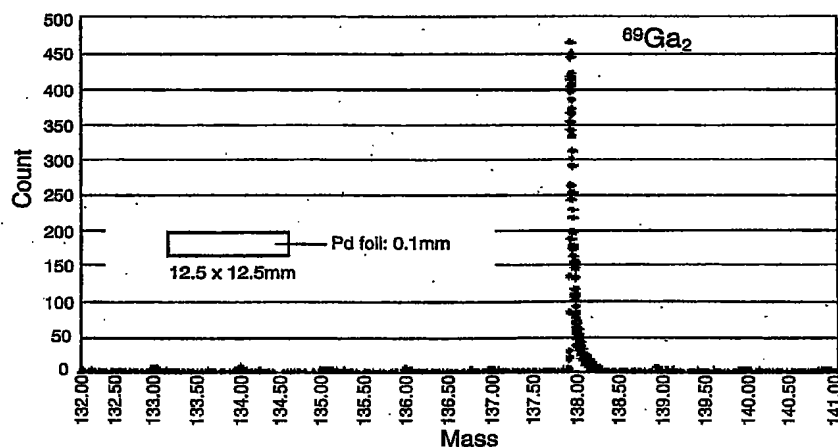


Figure 5. TOF-SIMS spectrum of mass number range 132–141 after deuterium permeation at 70°C for plain Pd foil.

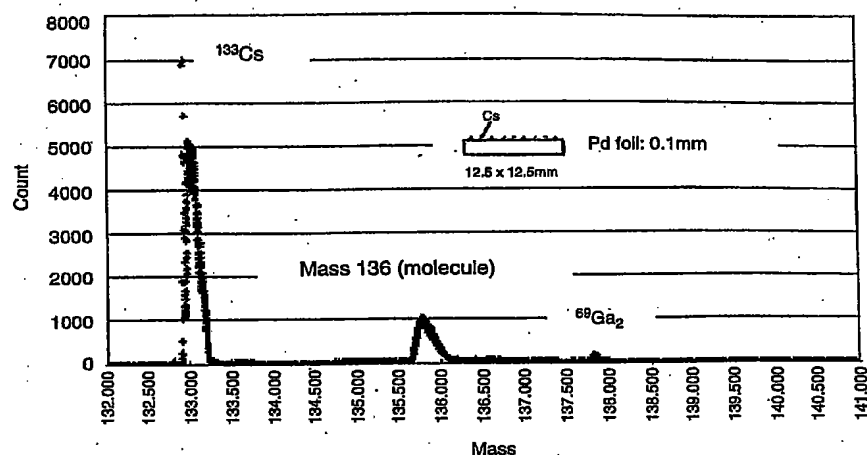


Figure 6. TOF-SIMS spectrum of mass number range 132-141 after deuterium permeation at 70°C for Cs deposited on plain Pd foil.

marked count except $^{69}\text{Ga}_2$ was detected in this mass number range. A TOF-SIMS spectrum of mass number range 132-141 after deuterium permeation at 70°C for Cs deposited on plain Pd foil is shown in Fig. 6. Three peaks at mass number 133, 136, and 138 can be seen in the spectra; these count peaks correspond to Cs, a molecule and Ga_2 , respectively.

TOF-SIMS spectra of the multi-layered sample with small amount of Cs after deuterium permeation at 70°C are shown in Figs. 7-11. Figure 7 shows a spectrum

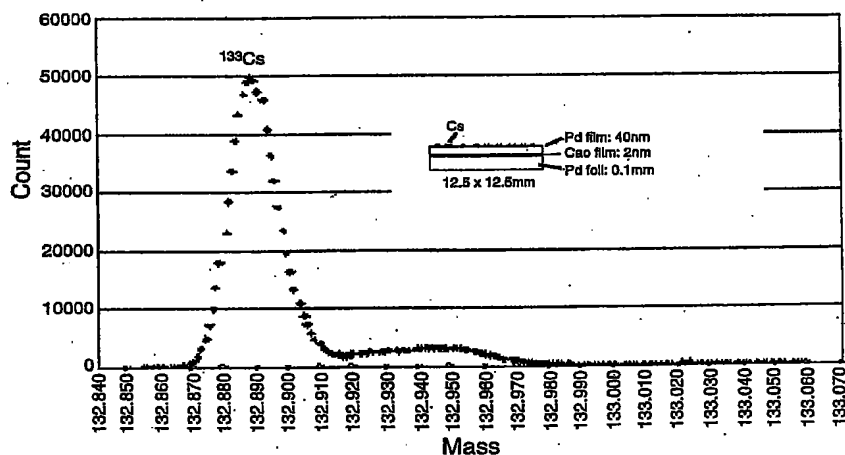


Figure 7. TOF-SIMS spectrum around mass number 133 after deuterium permeation at 70°C for Cs deposited on multi-layered Pd sample.

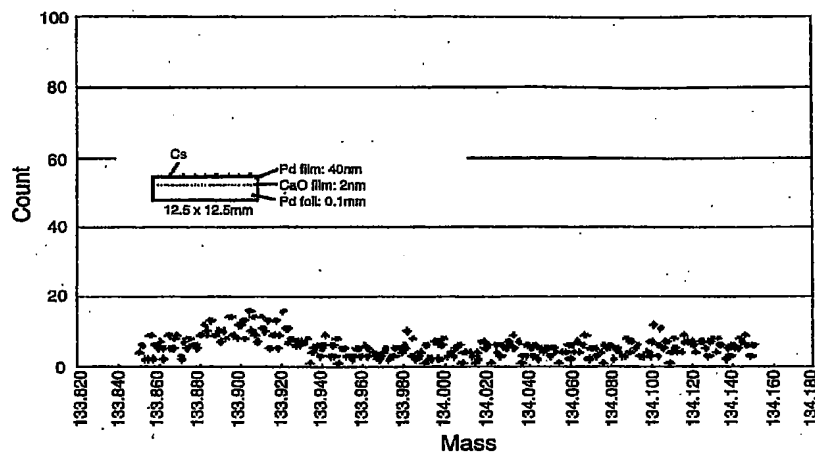


Figure 8. TOF-SIMS spectrum around mass number 134 after deuterium permeation at 70°C for Cs deposited on multi-layered Pd sample.

around mass number 133, where only the counts corresponding to Cs is seen at mass number 133. Figures 8 and 9 show spectra around mass number 134 and 141, respectively. No marked count was observed in both the spectra. It was not likely that the element ^{141}Pr was produced using the multi-layered sample prepared. However, one can recognize a small peak at mass number 141; which may imply a very small amount of Pr production.

To the contrary, we have found anomalous peaks at mass number 135 and 137, as shown in Figs. 10 and 11, respectively. The substance, corresponding to mass

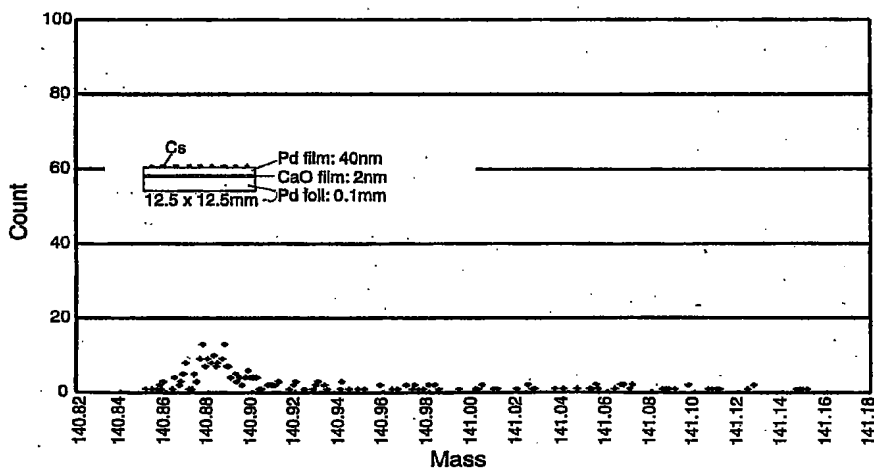


Figure 9. TOF-SIMS spectrum around mass number 141 after deuterium permeation at 70°C for Cs deposited on multi-layered Pd sample.

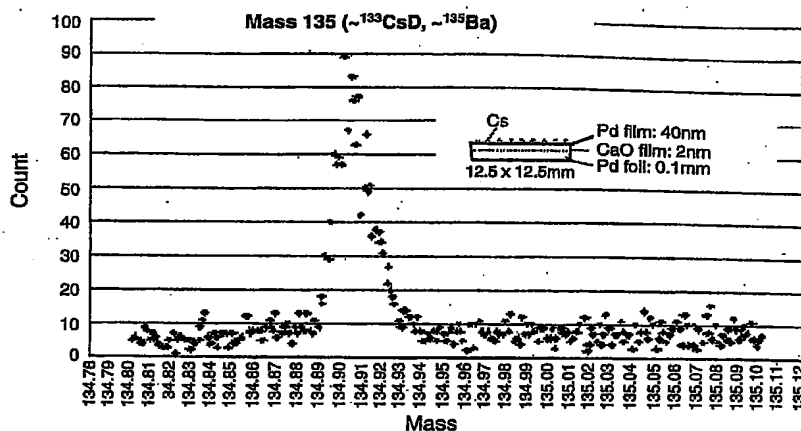


Figure 10. TOF-SIMS spectrum around mass number 135 after deuterium permeation at 70°C for Cs deposited on multi-layered Pd sample.

number 136, is considered to be a molecule. A small peak at mass number 135 may correspond to molecule CsD or an element such as ^{135}Ba . It should be noticed that the count intensity seen at mass number 137 is higher than that at mass number 135. This is the characteristic commonly observed in all the three measured areas. In general, amount of CsD formed is much more than that of CsD_2 during the deuterium permeation experiment. Accordingly, the substance corresponding mass number 137 could not be CsD_2 . While, the compounds consisting of Pd, which

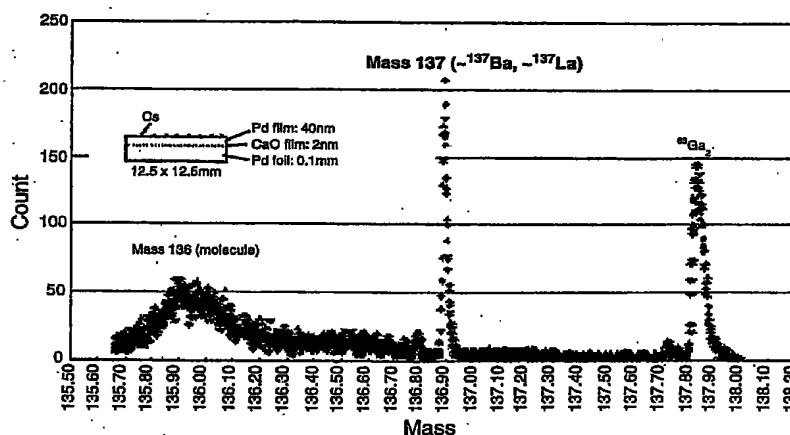


Figure 11. TOF-SIMS spectrum around mass number 137 after deuterium permeation at 70°C for Cs deposited on multi-layered Pd sample.

might be formed during the experiment, cannot account for these anomalous counts at mass number 135 and 137. Because no peak appeared at mass numbers 134 and 138 even though Pd has large natural isotopic ratios in mass number 105, 106, and 108.

The candidate elements and molecules detected by TOF-SIMS in mass number range 133–141 are compiled into Table 1. Most of all the sections are blank. This means almost no count was detected at each mass number. Since no substance with mass number 137 was registered in a control multi-layered Pd sample and in a plain Pd sample with Cs after the permeation, the process of forming multi-layered sample and of depositing Cs seemed to present almost no amount of contaminant of mass number 137 to these samples. Thus, the substance with mass number 137 observed after deuterium permeation is unlikely to be contaminant but would be an element such as ^{137}La or ^{137}Ba . The results suggest an impotent role of alpha cluster in the transmutation process of 8 and 12 mass number increasing.

Table 1. Candidate elements and molecules detected by TOF-SIMS at mass number range 133–141; blank means almost no count being observed

Mass	Plain Pd	Control Pd + Cs	After permeation Pd + Cs	Control Pd + CaO + Pd + Cs	After permeation Pd + CaO + Pd + Cs
133		Cs	Cs	Cs	Cs
134					
135					
136			Molecule	Molecule	Ba CsD
137					Molecule
138					Ba La
141					

4. Conclusion

Elements analysis on the Pd samples was performed after deuterium permeation experiment and for control Pd samples using TOF-SIMS. The TOF-SIMS has provided the marked count peaks at mass numbers 135 and 137 in spectra after deuterium permeation at 70°C, only when the multi-layered Pd sample with a small amount of Cs was used. The substance with mass number 137 could be ^{137}La or ^{137}Ba produced during deuterium permeation by some nuclear transmutation occurring on/in the uppermost of multi-layered Pd sample. The single couple of Pd/CaO thin films on Pd foil might contribute to induce production of an element with mass number 137. This would imply a transmutation of 4 mass number increasing before ^{141}Pr production.

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